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SOME CHEMICAL PROPERTIES OF EUCALYPTUS IN RELATION TO THEIR EVOLUTIONARY STATUS

James B. McNair

This paper presents comparisons of morphological and chemical characteristics of members of the genus *Eucalyptus* and shows that primitive species are primitive both morphologically and chemically and more recent species are advanced both morphologically and chemically. Chemical advance involves oxidation. But chemical and morphological advances do not necessarily progress hand-in-hand.

Baker and Smith (1, 2) after thirty years work classified the genus chemically and phylogenetically. Their phylogenetic ar-

GROUP I
PINENE $C_{10}H_{16}$
CINEOLE $C_{10}H_{18}O$, 4%
(FEATHER VENATION)

GROUP II
PINENE $C_{10}H_{16}$
CINEOLE $C_{10}H_{18}O$, 25%

GROUP IIIa, IIIb
PINENE $C_{10}H_{16}$
CINEOLE $C_{10}H_{18}O$, 49-64%

BLOODWOODS

STRINGY BARKS

IRONBARKS

GROUP IVa, IVb
PINENE LESS
CINEOLE 56-46%
PHELLANDRENE $C_{10}H_{16}$
AROMADENDRAL $\begin{cases} C_{10}H_{16}O \\ C_{10}H_{16}O \end{cases}$

GROUP V
PINENE
CINEOLE 16%
PHELLANDRENE

GROUP VI
PINENE
CINEOLE 13%
PHELLANDRENE

GROUP VIIa, VIIb
CINEOLE 15-3%
PHELLANDRENE
PIPERITONE $C_{10}H_{16}O$
(BUTTERFLYWING VENATION)
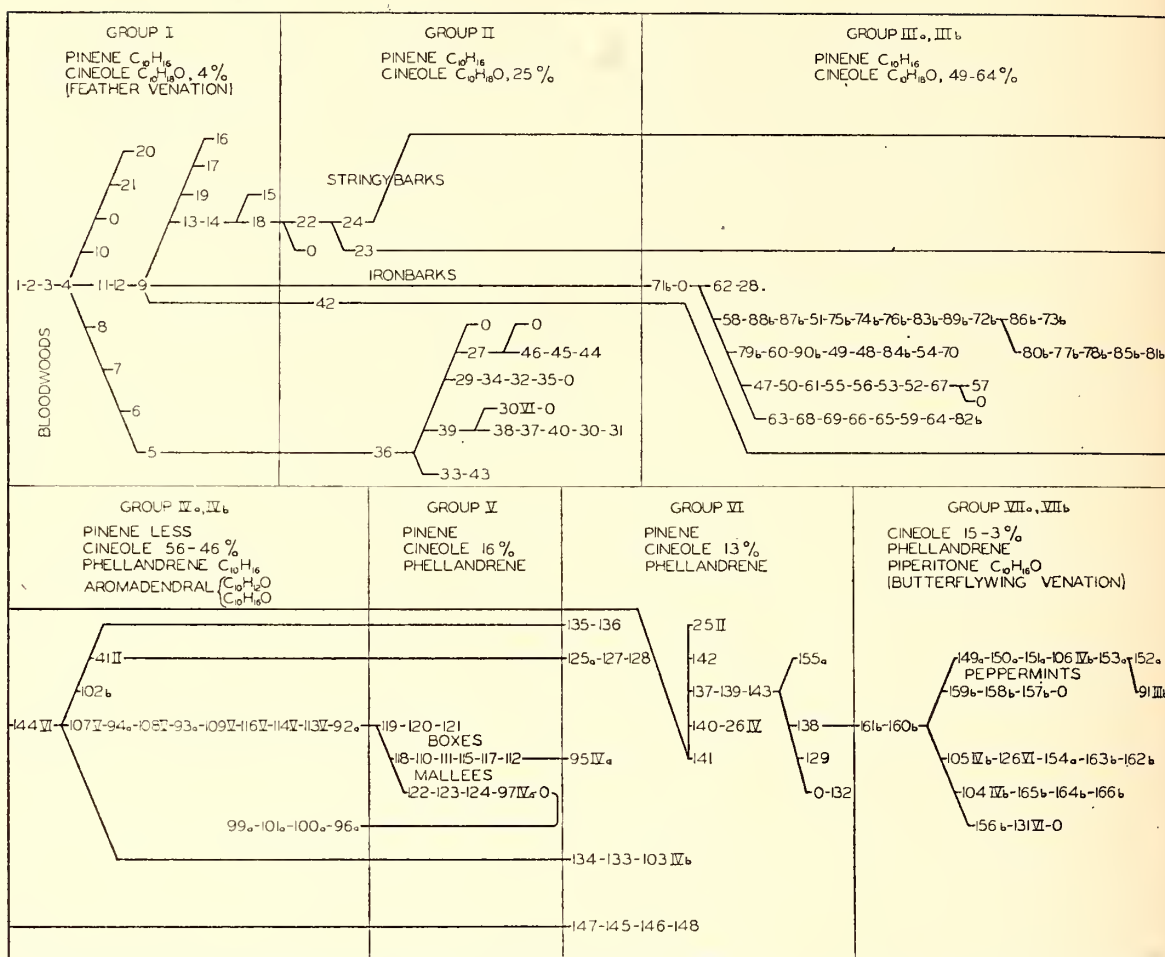
BOXES

MALLEES

PEPPERMINTS

Fig. 1. Phylogenetic tree of *Eucalyptus* species portraying both systematic and chemical relationships.

rangement is shown in figure 1 and their seven chemical groups are given in Table 1. If we consider the groups of Baker and Smith in relation to the phylogenetic tree made by them it is apparent that each group represents a horizontal cross section of their phylogenetic tree (fig. 1) and consequently sometimes includes heterogeneous species which are products of different branches of descent (not natural groups or genetic sequences) and sometimes includes reversions. In other words each Baker and Smith group represents a stratum or phase in the evolutionary development of the genus. So we find (fig. 1) that group II includes three separate natural sequences, 22. *E. Wilkinsonia,* 23. *E. eugenioides,* and 24. *E. umbra* all descendants of 18. *E. nigra;* 42. *E. peniculata* from 9. *E. botryoides;* and 36. *E. maculata* and its numerous following from *E. terminalis.* Group VI has five separate systematic series and in groups IVa and IVb are found examples of natural sequences on a morphological basis which include a variety of interspersed chemical groups. For example 144. *E. obliqua* of group VI chemically is followed morphologically by 107. *E. tereticornis* of group V chemically, followed by 94. *E. tereticornis* var. *cineolofera* of group IV class (a) chemically, etc.

Consequently it is apparent from figure 1 that these plants

may sometimes combine advanced systematic characters with more primitive chemical characteristics, for example, 105 IVb which is chemically in group IV but morphologically in group VII. In still other cases primitive morphological characters seem to have been retained by plants in chemically advanced positions, for example, 114 V between 116 V and 113 V. It may be concluded, therefore, that the development of chemical characteristics and morphological characters do not necessarily proceed hand-in-hand; one may proceed more rapidly or less rapidly than the other. An excellent example of this is seen in the case of *E. dives, E. radiata* and *E. micrantha.*

Eucalyptus dives, the common broad leaf peppermint, occurs in Australia over vast areas and the oil from the leaves has become of importance owing to the occurrence in it of from 40 to 50 per cent of the ketone piperitone, a commercial source of thymol and menthol. With the increased economic demand it was found that certain oils said to be obtained from the leaves of *E. dives* only yielded 5 to 20 per cent of the ketone. These were at first regarded as adulterated. It was, however, shown that they were genuine oils and that *E. dives* existed in at least four varieties which were morphologically absolutely indistinguishable both in the field or in the herbarium. At first sight this difference might be expected to be due to soil or climatic conditions but this is not the case, since the different varieties may grow side by side in the field and breed true when grown in pots (7, 8, 9, 10). The young seedlings from all four kinds were morphologically identical, yet when tiny fragments of the leaves, even from plants only one and one-half inches in height were rubbed between the fingers the characteristic odor of each particular kind was readily detected.

As a result of investigations by Penfold and Morrison (11, 12, 13) extending over a period of ten years it has been found possible to separate morphologically identical trees of *E. dives, E. radiata* and *E. micrantha* into groups as a result of hybridization based upon the chemical composition of the volatile oils.

Interrelationships Between Principal Constituents

The principle constituents of *Eucalyptus* oils are the two terpenes, pinene and phellandrene, the oxide cineole, the group of aldehydes and a ketone known collectively as aromadendral and the ketone piperitone.

Pinene. The pinene occurring in *Eucalyptus* oils is alpha pinene and it is found in two forms, one which deflects a ray of polarized light to the right (dextro) and the other which deflects to the left (laevo).

Phellandrene. This terpene is somewhat extensively distributed in the oils of certain groups of *Eucalypti*. It is more pronounced in those belonging to the more recent end of the genus, and it occurs more abundantly in those species common to the

Table 1. Amounts of Principal Substances and Physical and Chemical Properties of Eucalyptus Oil Groups (Australian and Tasmanian)

	Group No.	Pinene $C_{10}H_{16}$	Cineole $C_{10}H_{18}O$ percentage	Phellandrene $C_{10}H_{16}$	Aromadendral Cuminal $C_{10}H_{12}O$ Cryptal $C_{10}H_{16}O$	Piperitone $C_{10}H_{16}O$	Refractive index 20° C.	Specific gravity 15° C.	Optical activity	Solubility in parts 70–80 per cent alcohol, average
Large Group No. 1	I	large	rare 4.2	absent	......	......	1.4755	0.8863	+ 13.4	7.8
	II	large	40 – aver. 25	absent	absent	......	1.4705	.9015	+ 5.56	3.6
	III(a)	large	40–50 aver. 49	absent	rare	......	1.4670	.9159	+ 6.1	2.3
	III(b)	large	55 + aver. 64	absent	absent	......	1.4656	.9178	+ 4.28	1.4
	IV(a)	diminishing	40 + aver. 56	absent	present	......	1.4677	.9179	+ 0.5	2.6
	IV(b)	......	40 + aver. 46	present	......	......	1.4682	.9119	– 4.5	
Large Group No. 2	V	present	40 – aver. 16	rare	present	......	1.4800	.9056	– 4.0	2.3
	VI	present	40 – aver. 13	present	......	......	1.4845	.9054	– 7.0	2.1
	VII(a)	......	40 – aver. 15	large	......	present	1.4793	.8863	–32.7	2.0
	VII(b)	......	rare aver. 3	present	......	present	1.4838	.8803	–38.8	

eastern and southeastern portion of the continent of Australia and Tasmania.

CINEOLE. The constitution of cineole is probably that of cinyl oxide, $C_{10}H_{18}O$. It has been described under the names eucalyptol (its most common commercial name) and cajuputol.

When terpin is dehydrated cineole is produced. Cineole is an internal ether produced by the elimination of water between the two hydroxyl groups in terpin. Although we have no direct evidence as to the mechanism of the formation of cineole in plants, its very frequent occurrence in oils containing α-terpinene can be accounted for readily if it be assumed to be formed from either α-pinene or α-terpineol, when terpin may be regarded as an intermediate product (16, 382).

The oils from the group of *Eucalypti* known as "gums" usually contain a fairly large amount of cineole, together with pinene, and in the case of many members belonging to this group, the cineole increases in amount when the oils are stored. This increase in cineole through possible oxidation during storage apparently confirms the fact that cineole is an oxidation product of pinene.

AROMADENDRAL. The term aromadendral is used to denote the presence of one or more members of a group of characteristic aldehydes and a ketone in *Eucalyptus* oils. These aldehydes include cuminaldehyde (cuminal), phellandral and cryptal; they do not seem to occur in the oils of the earlier members of the genus (the pinene yielding group), nor in those of the more recent species, particularly those in which phellandrene is the more pronounced terpene. In these latter species the characteristic constituent is the ketone piperitone.

It may also be stated as a general rule that cymene is present in either larger or smaller amount in the oils of species containing these aldehydes.

Wallach (17) has shown that the oxidation of β-phellandrene produced a glycol, which on treating with dilute H_2SO_4 gives dihydro- and tetrahydrocuminaldehydes. Molecular re-arrangement of a somewhat similar character may perhaps take place naturally.

PIPERITONE. Piperitone is the peppermint ketone of *Eucalyptus* oils. It is an unsaturated ketone $C_{10}H_{16}O$ with one double bond. It appears to occur only in the oils of species occupying the more recent end of the genus and is not found in the oil of any member of the groups occupying the anterior position, in the evolutionary sequence of the genus. In the oils of most species, piperitone is found associated with the corresponding l-rotatory secondary alcohol piperitol. Phellandrene in *Eucalyptus* oils is often associated with l-piperitone, but not always, although it may be accepted that the most pronounced phellandrene *Eucalyptus* oils always contain this ketone in smaller or larger amounts.

Hughesdon, Smith and Read (4) have directed attention to the fact that l-piperitone always occurs in nature in association with
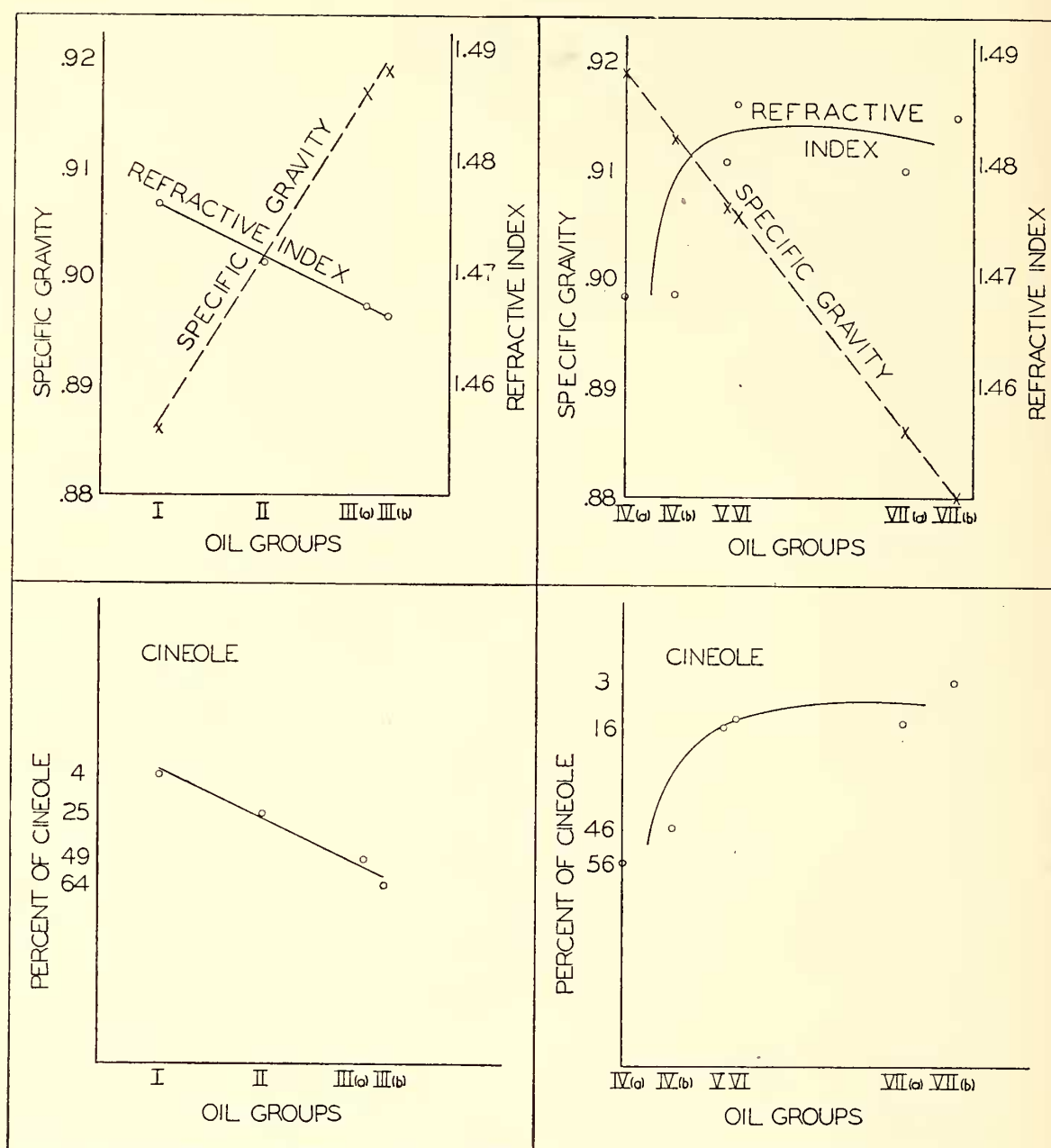
Fig. 2. Graphs showing the specific gravities, refractive indices and percentages of cineole of *Eucalyptus* oil groups.

l-α-phellandrene and l-piperitol and it is possible that some relationship exists between these substances. In the laboratory it has been found that the reduction of either d- l- or dl-piperitone with sodium in alcoholic solution yields dl-isomenthols and dl-menthols, some dl-α-phellandrene being formed simultaneously.

Comparison of Specific Gravities and Refractive Indices

Based on their specific gravities (and concomitant refractive indices) *Eucalyptus* oils may be divided into two large groups. The first and most primitive group includes groups I, II, IIIa and IIIb of Baker and Smith (2).   (Table I and text fig. 2).

The second and less primitive group includes groups IVa, IVb, V, VI, VIIa and VIIb of Baker and Smith (2).

In figure 2 it so happens that a straight line will pass near all

the points representing the specific gravities of groups I to IIIb inclusive and that another straight line will pass near all the points representing refractive indices.   Therefore, the rule that a high specific gravity is accompanied by a low refractive index and vice versa for plant volatile oils (5) is confirmed.

It is noticeable that a straight line cannot be drawn through all of the five points representing the refractive indices of groups IV to VIIb, and that the line of closest fit is a curve.   Consequently it is obvious that the mixture of substances which constitutes the volatile oils of at least some of the members of this group are different from those of the groups I to IIIb where straight lines can be drawn approximately through all the points representing the specific gravities and another straight line will lie near to all of the points representing refractive indices.   Different mixtures of substances are also indicated in figure 2 by the fact that for groups I to IIIb the refractive index decreases with advance in plant evolutionary position while for groups IV to VII the reverse is true.   Likewise there is a difference in the directional change in specific gravities for groups I to IIIb and IV to VII.   In groups I to IIIb the specific gravity increases with advance in plant evolutionary position while in groups IV to VII the reverse takes place.

INCREASE IN OXYGENATED BODIES WITH EVOLUTIONARY PROGRESS

According to the general theory for angiosperm volatile oil behavior, the specific gravity should increase and the refractive index should decrease with advance in evolution (6).   The oils of groups IVa to VIIb contain principally pinene, its oxidation product cineole, and phellandrene and its oxidation products, cuminal, cryptal, phellandrol and piperitone.   From Table 1 it is seen that pinene and cineole are diminishing while the other substances mentioned are increasing with progress from IVa to VIIb.   The removal of a heavy substance from the oil would decrease its specific gravity.   Therefore the subtraction of pinene which has a specific gravity of 0.865 at 15 degrees would not tend to decrease the specific gravity of the oil because its specific gravity is lower than that of the average specific gravity of groups IVa to VIIb (Table 1).   However, if cineole be taken out the specific gravity would tend to lower because cineole with its specific gravity of 0.930 at 15 degrees is greater than the average specific gravity of the oils of the groups here considered.   The content of cineole rapidly diminishes (56 per cent to 3 per cent) in these groups. The other main oxygenated bodies of these groups also have specific gravities near that of cineole.   Consequently neither the amounts of these compounds nor that of cineole increases from groups IVa to VIIb enough to change the diminishing trend of the specific gravity.   Should the total amount of the oxygenated bodies increase from IVa to VIIb it is obvious that the specific

gravity would also increase. Consequently we may conclude that the volatile oils of *Eucalyptus* species when in their native habitat, increase in specific gravity with the increase in plant evolution *except* where the oxygenated bodies decrease in amount with the increase in evolution.

The refractive index of cineole (1.4596 at 20 degrees) is lower than the average refractive index for the group oils. Consequently its removal would tend to raise the refractive index of the group oils. Otherwise the refractive index would increase with evolutionary progress.

Table 1 shows that chemical analysis has found the number and variety of oxidation products to increase with advance in evolutionary position in the genus. Cineole, the oxidation product of pinene, is found in all of the groups. Aromadendral (which is mostly cuminal and cryptal oxidation products of phellandrene) is found in groups III, IV and V. Piperitone with which is associated phellandrol, both oxidation products of phellandrene are found in group VII.

The increase in the number and variety of oxidation products is likewise shown by the increase in the solubility of the crude oils in 80 per cent alcohol from groups IVa to VIIb (Table 1). For the oxidation products are soluble in alcohol whereas the terpenes, pinene and phellandrene are comparatively insoluble. The amount of cineole rapidly decreases from group IVa to VIIb and yet the solubility increases. Consequently some other oxidation products must replace the removed cineole.

Optical Rotation

A glance at Table 1 shows clearly that the average optical rotation becomes more laevo-rotatory as the evolutionary sequence of the groups advance. There are a number of dextro- and laevo-rotatory substances in the oils. There are present in varying amounts both dextro- and laevo-pinene and d- and l-phellandrene. There are also l-piperitone, d-terpinol and d-eudesmol. Most of the pinene is dextro- and most of the phellandrene is laevo-rotatory; d-pinene accounts for the predominant d-rotation in groups I, II, IIIa and IIIb and l-phellandrene and l-piperitone for the predominant l-rotation in the remaining groups.

Ontogeny and Phylogeny

The *Eucalypti* furnish an excellent chemical counterpart to the morphological theory of Haeckel (3) that ontogeny recapitulates phylogeny, that the organism in its development is to a great extent an epitome of the form modifications undergone by the successive ancestors of the species in the course of their historic development. Oil from the younger seedlings contains more d-pinene and less cineole (that is more hydrocarbon and less oxygenated products) than does that from the saplings two or three

years old, and the maximum cineole content is reached in the oil collected from older trees (2). This is true also for the leaves which are reproduced from lopped old trees, and the oil from seven months "suckers" contains more cineole and less pinene than does that from twelve months old seedlings, while that from fifteen months old "suckers" follows the same rule in respect to two and one-half year old seedlings. The increase in the amount of cineole with progress in evolution in Large Group No. 1 of the genus is shown in Table 1.

SUMMARY

There has been orderly evolution in volatile oil characteristics of the *Eucalypti* which may be correlated with changes in morphological characteristics. However, oil groups represent cross-sections of the phylogenetic tree and not necessarily genetic sequences or natural groups of the systematist. Botanically distinct species are generally distinguished by their chemical constituents and also in some cases where morphological examination shows little or no difference, chemical analysis of the oil reveals the existence of completely distinct varieties.

With progress in evolution the amount of the hydrocarbon (terpene), pinene and its oxidation product cineole increase to a maximum, then decrease; another hydrocarbon (terpene) phellandrene and its products form a second series of compounds which is evidenced first by the appearance of aromadendral (an aldehyde and ketone mixture containing cuminaldehyde and cryptal); next phellandrene becomes a pronounced constituent and finally its ketone piperitone increases in amount.

The specific gravity increases with the increase in plant evolution, and the refractive index decreases with the increase in plant evolution, except where the oxygenated bodies decrease in amount with the increase in evolution.

The number and variety of oxidation products increase with advance in evolutionary position in the genus.

There is a tendency for the optical rotation to become more laevo-rotatory with advance in evolution caused principally by a decrease in d-pinene and an increase in l-phellandrene and its ketone l-piperitone.

Morphological and chemical phylogeny have their counterparts in ontogeny.

Los Angeles, California,
July 3, 1941.

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GRASSLAND AND RELATED VEGETATION IN NORTHERN MEXICO

Forrest Shreve

The extensive grassland area of the central United States exhibits its optimum development in Kansas and Nebraska, and extends south to the Mexican boundary only after suffering localization in occurrence and modification in character. The plains and gently falling outwash slopes of southern New Mexico and western Texas are largely occupied by a very open type of arid grassland in which *Yucca, Nolina, Dasylirion, Agave, Opuntia* and various shrubs are conspicuous. This is a transition region, in which the conditions are intermediate between the optimum ones for grassland and for desert. The vegetation is formed by an infiltration of plants from each of these vegetations, with very few dominant species that are distinctive of the transition region. In both of the states mentioned and also in southeastern Arizona there are areas of true grassland growing in favorable valleys or